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An adaptive immune optimization algorithm with dynamic lattice searching operation for fast optimization of atomic clusters

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ABSTRACT

Geometrical optimization of atomic clusters is performed by a development of adaptive immune optimization algorithm (AIOA) with dynamic lattice searching (DLS) operation (AIOA-DLS method). By a cycle of construction and searching of the dynamic lattice (DL), DLS algorithm rapidly makes the clusters more regular and greatly reduces the potential energy. DLS can thus be used as an operation acting on the new individuals after mutation operation in AIOA to improve the performance of the AIOA. The AIOA-DLS method combines the merit of evolutionary algorithm and idea of dynamic lattice. The performance of the proposed method is investigated in the optimization of Lennard-Jones clusters within 250 atoms and silver clusters described by many-body Gupta potential within 150 atoms. Results reported in the literature are reproduced, and the motif of Ag_{61} cluster is found to be stacking-fault face-centered cubic, whose energy is lower than that of previously obtained icosahedron.

1. Introduction

Atomic clusters have long been studied by chemists and physicists because of the extremely important and unique properties [1]. For instance, Ag_3 clusters and ~3.5-nm Ag nanoparticles might provide highly efficient catalysts for propylene epoxidation [2]. Pt_{8-10} clusters stabilized on high-surface-area supports are 40–100 times more active for the oxidative dehydrogenation of propane than the corresponding platinum and vanadia catalysts [3]. Such properties of clusters are generally dependent upon their geometrical structures, which have been widely studied both theoretically and experimentally. To determine the global minimum configurations of clusters, molecular mechanics (MM) [4], molecular dynamics (MD) [5], quantum mechanics (QM) [6], and the mixed QM/MM methods were usually applied [7,8]. QM can accurately determine the energy of clusters based on the calculations of the electronic structures, but the related calculation is usually very time-consuming with respect to powerful computational resources. However, because the electronic structure is ignored in MM, the properties of large-size clusters can be investigated. Recently, global optimization method combined with density functional theory (DFT) has been used to perform global optimization of B_{14}^{2+} cluster [9], $(TiO_2)_n$ (n = 1–6) clusters [10], and 8-atom Au–Ag nanoalloys [11]. Therefore, considering the complexity of computation, the efficiency of optimization algorithms is very important for the determination of structures for clusters.

Geometrical optimization of atomic clusters belongs to the class of NP-hard (nondeterministic polynomial time) problem, and lots of efficient algorithms have been developed for the problem [12]. These optimization techniques can be divided into three classes. The first class belongs to evolutional algorithm, and the representative examples include the genetic algorithm (GA) [12–14], fast annealing evolutionary algorithm (FAEA) [15], particle swarm optimization (PSO) [16], artificial bee colony (ABC) [17], adaptive immune optimization algorithm (AIOA) [18,19], and population-based algorithm [20]. The basic frame of algorithms is almost similar, e.g., including the generation of initial configurations, individual selection, and immune operation. Besides for atomic clusters, they were also successfully used in other areas, e.g., for a quantitative production of the Butyrylcholinesterase inhibitors with 3D-QSAR [21] and nonlinear fitting of powder EPR spectra [22]. The second class algorithm has proved to be capable of overcoming large energetic landscape by transforming the potential energy surface, such as basin-hopping (BH) method and its related variants [4,23], minima hopping [24], self-consistent basin-to-deformed-basin mapping [25], and funnel hopping method [26]. The third class is related to a technique called dynamic lattice (DL) [27]. By moving the active atom with the highest energy to the vacant lattice site with the lowest energy, the possible location sites of the starting local minimum can be found from the constructed DLs. The proposed dynamic lattice searching (DLS)

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algorithm runs with a high efficiency [27]. Improved algorithms based on DLS include DLS with constructed core (DLSc) [28], DLS with interior operation (DLS-IO) [29], DLS with rotation operation (DLS-RO) [30], and DLS with two-phase local minimization method [31]. The method is also applied as surface operation in heuristic algorithm combined with the surface and interior operators (HA-SIO) [32]. Other efficient algorithms include big-bang method [33], and continuous extremal optimization (CEO) [34].

To complete the above methods, various operations have played an important role for cluster geometry optimization problems. In these operations, cut and splice like crossovers are the effective recombination operators [14]. Recently, it was reported that the algorithm with the sphere-cut-splice crossover exhibited better performance than the one with the plane-cut-splice crossover [35]. In GA, mutation operation was performed to avoid stagnation and to maintain population diversity, and a number of mutation schemes, such as atom displacement, twisting, cluster replacement, and atom permutation, were adopted. A rotation operation was designed to realize the structural transformation from decahedra to icosahedra for the optimization of large clusters, by rotating the atoms below the center atom with a definite degree around the fivefold axis [30]. Moreover, the use of approximate symmetry substantially improved the efficiency of global optimization for the atomic clusters [36].

AIOA adopts the basic frame of evolutionary algorithms, such as GA, and its high performance has been proved. In AIOA, a cluster similarity checking method with the connectivity table was utilized to keep diversity of the population, and the energy-based mutation operation helped to increase population diversity by moving atoms with higher energy to a random location on the surface of the cluster. However, for a robust method, the mutation operator is insufficient for getting the optimal motifs. Moreover, the technique of DLS could rapidly make the clusters more regular and greatly reduces the potential energy. DLS can thus be used as an operation acting on the new individuals after mutation operation in AIOA to improve the performance of the AIOA. Therefore, based on the fast DLS, an improved AIOA, called AIOA-DLS, is proposed in this work, which combines the merit of evolutionary algorithm and idea of dynamic lattice. The performance of AIOA-DLS is studied in the optimization of Lennard-Jones $(LJ)_N$ ($N = 13$–250) clusters and Ag_N ($N \leqslant 150$) with Gupta potential. Moreover, a new global minimum with stacking-fault fcc motif of Ag_{61} cluster is found.

2. Method

2.1. Dynamic lattice searching operation

The basic idea of the DLS is that just specific positions could be located after local minimization (LM) when one atom is added to a fixed cluster. All possible locations surrounding an unknown local minimum correspond to DL, and the run of DLS is related to the DL construction and DL searching. It can be summarized as a starting local minimization step, and the repetition of DL construction and DL searching. The detail steps of the DLS are as following:

(1) Randomly generate an initial configuration s with N atoms, and it is then local minimized with the run of the limited-memory quasi-Newton method (L-BFGS) [37].
(2) Perform DL construction. At first, the coordination of the configuration s is centered, and the maximum radius of the cluster R_{max} is obtained. Then, the nth shell of the Mackay icosahedron is used to simulate a uniform distribution on the surface of the configuration s, and its radius is $R_0 = R_{max} + 0.2$. These N_{PL} sites are the primary lattice (PL) sites. It can be found that N_{PL} is a parameter determined by cluster sizes. Next, an atom is added to each site of the PL, and sub-LM is operated on the atom to generate a new position. If a pair of positions are too close, i.e., the distance is smaller than 0.05, they are considered as one position. Based on PL, N_{DL} DL is constructed. In addition, the selected N_{mov} active atoms, i.e., the atoms with highest sub-energy in the current cluster, are merged into DL. Finally, the total size of the DL sites is thus $N_{DL}' = N_{DL} + N_{mov}$.
(3) DL searching procedure. The energy of a cluster may be largely decided by N_{mov} atoms with highest sub-energies. Therefore, the task of DL searching is to find the possible combination of N_{mov} atoms from the current DL, which becomes a combinational optimization problem. The simple greedy method (SGM) is utilized to solve the problem, and a single SGM can be summarized as: (i) Randomly select N_{mov} sites (occupied sites) from DL to form an initial solution S_k. (ii) Move the atom located at the occupied site with highest energy to the unoccupied site with lowest energy in the DL to generate a new solution S_k'. (iii) If $(E(S_k') < E(S_k)$, take S_k' as the starting solution of the next generation (S_{k+1}) and return to step ii. Otherwise, S_k is taken as the best solution of this SGM run. Actually, one run of SGM cannot guarantee the success, so N_{try} SGM is calculated. N_{best} solutions with lower energies are kept for further LM, and the solution s' with the lowest energy will be selected as the final solution of this DL searching.
(4) If $E(s') < E(s)$, s' is selected as the starting cluster, and return to step (2). Otherwise, solution s is the result of the DLS run.

It has been proved that DLS is a high efficient algorithm for geometrical optimization of clusters, e.g., Lennard-Jones (LJ) clusters [27], Ag clusters [38], Al clusters [39], C_{60} clusters [40]. It should be noted that any configuration can be further optimized by the DLS run, so it may be applied as an operation in other algorithms to decrease potential energy rapidly.

2.2. Description of the AIOA-DLS algorithm

The AIOA-DLS procedure displayed in Fig. 1 can be described as following:

(1) Randomly generate a certain number (N_{lib}) of initial configurations in a sphere with a radius of $R = (N/17.77)^{1/3} + 1.0$, where N is the size of cluster. Then these configurations are locally optimized. At $it = 0$, initial gene library is formed by these configurations.
(2) At the itth generation, the fitness (fit(v)) and density ($C(v)$) of the individual v in the gene library are calculated, and the details of the calculation were described in Ref. [18].
(3) The selection probability e_v of individual v is determined by the fitness and density: $e_v = \text{fit}(v)/C(v)$. Then N_{pop} individuals are selected from the gene library by the immune selection procedure.
(4) Energy-based mutation operation is performed on the selected individuals. This operation is based on the fact that atoms with lower connectivity number (nn) will have higher potential energy, and the mutation rate of an atom in a cluster is with respect to nn. While an atom is selected, it will be randomly moved to a radius of r_0 (equilibrium distance between two atoms). The mutated individuals are then minimized with the LM procedure.
(5) DLS operation is operated on the individual v after mutation operation. Note that the starting configuration of the DLS is the individual v, and the optimized individual is w.